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Sputtering and surface topography modification of bismuth thin films under swift $^{84}\mathrm{Kr}^{15+}$ ion irradiation

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ABSTRACT

The sputtering and surface topography modification of bismuth thin films deposited onto Si substrates and irradiated by 27.5 MeV $^{84}\mathrm{Kr}^{15+}$ ions over the fluence range 10^{12}–10^{14} cm^{-2} have been studied using three complementary techniques: Rutherford backscattering spectrometry (RBS), atomic force microscopy (AFM) and X-ray diffraction (XRD). The RBS analysis reveals a linear reduction of the initial thickness of the irradiated bismuth samples by ~4% up to 7% with increasing ion fluence corresponding to a mean sputtering yield of $\sim 2.9 \times 10^2$ at/ion. Besides, significant sample surface topography changes occur upon ion irradiation consisting in grain growth and surface roughening clearly pointed out by performed AFM and XRD analyses. Moreover, a close correlation is observed between the variations versus ion fluence of the measured sputtering yield and the determined Bi surface grain size and compressive strain. These moderate Bi surface effects are similar to those pointed out previously for thin films irradiated by MeV heavy ions. They can be mainly caused by inelastic electronic collision mechanisms taking place within the Bi material electronic stopping power regime below the threshold for latent track formation.

1. Introduction

While traversing solid materials, energetic ions may undergo large energy losses and are consequently slowed down either via elastic nuclear collisions (for low energy incident ions in the keV range) or inelastic electronic collisions (i.e., excitations and ionizations of target atoms for high energy incident ions in the MeV–GeV ranges). Both two types of collisions give rise to damage and structure modifications in the solid often accompanied by the erosion of the target material surface (or sputtering). In the case of swift heavy ions (SHI), typically moving on straight paths in the target, material damage ranging from point defects up to phase transitions involving extended ion tracks of nearly cylindrical shapes are produced in insulators. In contrast, in pure metals the deposited electronic excitation energy is rapidly dissipated through the lattice before any damage can be created. From a theoretical point of view, unlike the processes of the initial kinetic energy transfer to target atoms, the mechanisms behind the conversion of the deposited electronic excitation energy into heat of the material lattice, then to complex atomic motions (atomic displacements, ion/atom collision cascades) inducing various types of material damage are yet far from being well elucidated. The nuclear sputtering of the material, induced by keV ions and presumably due mainly to elastic collision cascades of recoil target atoms, has been intensively investigated experimentally, by theory and by modeling during several decades [1–3]. The electronic sputtering, induced by SHI irradiation and expected to be caused by inelastic collisions, has also been studied principally for insulators and for some metallic targets [4–6] (and references therein). More than for other materials, in the case of semi-metallic (polycrystalline) targets having specific properties like bismuth (Bi), however, SHI irradiation-induced effects and underlying mechanisms are still much less investigated and well understood [7–10]. Target material damage induced by strong electronic energy losses under SHI irradiation has been evidenced experimentally in in-situ electrical resistance measurements for bulk Bi [8–10]. Two main distinct theoretical models of the electronic excitation energy transfer to the lattice atoms are usually invoked for describing the SHI irradiation-induced damage, which are: (i) Coulomb explosion [11] and (ii) thermal spikes mechanism [9], beside molecular dynamic numerical simulations [12]. According to the former model, valid at least at the initial stage of the material damage, a lattice zone around the incident ion central path is partially or fully stripped of electrons with the produced ions or bare nuclei being violently ejected radially outward under the action of strong electric forces. Then, they

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can be rapidly re-screened by initially freed target atomic electrons. In the second main electronic energy transfer model, excited electrons heat the lattice via electron–phonon coupling, which leads to a localized, transcient and highly disordered zone at high temperature, likely generated by nonlinear ion/atom collision cascades. Especially the latter so-called *thermal spikes* model was successfully used to account for SHI irradiation-induced surface damage such as the creation of latent tracks above a threshold value of the material electronic stopping power (~24 keV/nm for Bi, see reference [8]) leading to the amorphization of initially crystalline materials. For polycrystalline targets, like deposited Bi thin films, the mechanisms behind the sputtering process within the electronic stopping power regime should then operate differently regarding the crucial dependence of SHI irradiation-induced effects on target properties like the sample thickness and the grown grain size [4,13–16]. Obviously, the magnitude of the damage induced in the material also depends on the intrinsic properties of the latter (chemical bonding, electrical conductivity, aggregation state) and on SHI irradiation conditions like the incident ion energy and fluence. Therefore, materials of different natures are expected to be differently effected under the same ion irradiation conditions.

In the current paper, we report on a careful experimental investigation of the sputtering and surface topography modifications suffered by Bi thin films deposited onto Si substrates under normal impacts of 27.5 MeV ^{84}Kr^{15+} ions. Beside the main objective of performing quantitative measurements of the induced surface effects in Bi, we attempted to understand the experimental observations by highlighting, following other groups, the above ion-surface interaction mechanisms that are still being debated. For comparison to previous measurements and interpreting the observed (moderate) SHI irradiation effects, we will also refer to our recent works on similar Bi thin films irradiated by keV Ar$^+$ ions [17,18]. In those experiments, the sputtering yield data and other observed Bi surface effects were satisfactorily interpreted by primarily invoking interaction mechanisms supporting the generation of linear elastic collision cascades [2,19]. In the current study, evaluating Bi electronic and nuclear stopping powers (S_e and S_n) for 27.5 MeV Kr incident ions and the mean projected range (R_p) of the latter in Bi using the SRIM-2010 computer code [20], one finds values of ~8.08 keV/nm, 0.18 keV/nm and 5.35 µm, respectively. Thus, one has $S_e >> S_n$ and $R_p >> t \sim 0.3$ µm (the mean thickness of the Bi samples used in this experiment). Therefore, one expects all incident ^{84}Kr ions striking the Bi target film to fully cross the latter, stopping far away outside it. Notice that the estimated Bi stopping power is only of 1/3 of the threshold value for which one could assume strong SHI irradiation-induced surface damage to occur, like latent tracks or amorphization [8]. In these conditions, the observed modifications suffered by the Bi film surface upon SHI irradiation are moderate and likely essentially caused by electronic collision interaction mechanisms (ion/atom recoils and displacements). However, although one expects, in first sight, these processes to be the main cause behind the observed high Bi sputtering yield and other irradiation-induced surface effects, elastic nuclear collision mechanisms may also play a non negligible role in this respect.

In the following we, first, succinctly report (in Section 2) on the general experimental method, set up and techniques used described in more detail elsewhere [17,18]. Then, we present and discuss (in Section 3) the obtained experimental results before giving a summary and concluding (in Section 4).

2. Experimental

High purity (99.99%) thin Bi films were deposited by high vacuum evaporation (target chamber pressure of $\sim 8 \times 10^{-7}$ mbar) onto well cleaned and polished Si (100) substrates at room temperature using an electron beam. The evaporation rate during the film deposition process was kept between 2 and 4 Å/s for obtaining smooth sample surfaces [21]. The target sample thicknesses were estimated, first, by means of an *Inficon* monitor deposition apparatus connected to the evaporator displaying the deposited Bi layer with a relative uncertainty of ±1.5%; then, they were more quantitatively determined by RBS spectrometry. About 300 nm-thick Bi samples were used in these experiments. They were placed inside a scattering chamber under high vacuum (under mean pressure of $\sim 2 \times 10^{-6}$ mbar) and irradiated by 27.5 MeV ^{84}Kr^q ions at equilibrium charge state, $q = +15$, delivered by the 2nd solid-pole injector cyclotron (SPC2) of the iThemba Labs in Somerset West, South Africa. The irradiations were performed uniformly over each scanned Bi sample surface at room temperature with varying the incident ion fluence in the range 8.9×10^{12}–1.35×10^{14} cm^{-2} using a beam spot of rectangular shape (2.0 ± 0.1 cm × 1.5 ± 0.1 cm) that was incident normally to the Bi film surface. During SHI irradiations, the beam current intensity was maintained at a low mean value of ~107 nA (mean current density of $\sim 1.5 \times 10^{10}$ ions/cm^2/s) in order to avoid the excessive heating or melting of the samples [4]. Then, in a subsequent step, the irradiated samples were analyzed in thickness by RBS spectroscopy for evaluating the sputter yields (see Section 3 and Ref. [17]). The RBS analyses were conducted using a 2-MeV ^{4}He$^+$ ion beam delivered by the 5.5-MV Van de Graaff accelerator of the iThemba Labs with mean current intensity of ~40 nA. Backscattered alpha particles were detected by means of a well collimated surface barrier Si detector, placed 11 cm apart the Bi/Si target sample at 165° relative to the primary beam direction. The absolute uncertainties in the measured sputtering yields were estimated as in reference [17]. They essentially resulted from errors propagating in ion fluence expression (amounting to ~20%) and errors in the target sample areal densities (ranging only between 1% and 3%), and amount, at most, to ~25%. Finally, the AFM and XRD techniques (applied, respectively, at a 7° grazing incidence angle and in tapping mode) were used to investigate the surface topography and crystalline structure evolutions of the SHI irradiated Bi film surfaces.

3. Results, analyses and discussion

3.1. Sputtering yield, comparison to theory

Recorded RBS spectra for Bi/Si samples irradiated with incident 27.5 MeV ^{84}Kr^{15+} ions of fluences ranging between 8.9×10^{12} cm^{-2} and 1.35×10^{14} cm^{-2} are reported in Fig. 1, together with a reference spectrum for an un-irradiated sample. As observed in all such spectra, one can see that the initially deposited Bi film thickness significantly reduces upon SHI irradiation as ion fluence increases. One can also note the absence of any atomic interface mixing in the irradiated Bi/Si samples over the ion fluence range investigated. This observation is consistent with at least two previous results [22,23] for Bi/Si samples irradiated by SHI, suggesting that the effect of the latter on the target atomic mixing depends not only on the sensitivity of interface-forming materials to the electronic energy deposition but also on chemical driving forces that prevent inter-diffusion to occur in the thermal spike region. The RBS spectra for the irradiated samples were simulated by the RUMP computer code [24] in order to determine quantitatively the variation of the Bi sample thickness following SHI impacts. The areal density of the un-irradiated sample was estimated to be of $\sim(8.1 \pm 0.2) \times 10^{17}$ at/cm^2 corresponding to linear mean thickness, t, of ~289 ± 7 nm. However, as can be seen in Fig. 2, this initial mean value reduces linearly by ~4% up to 7% with increasing incident ion fluence. As proceeded in reference [17], the mean rate of removed Bi material